



1 **The conclusive impact of aerosols vertical structure on low-**
2 **atmosphere stability and its critical role in aerosol-PBL interaction**

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24 **Abstract.** Aerosol-planetary boundary layer (PBL) interaction was proposed as an
25 important mechanism to stabilize atmosphere and exacerbate surface air pollution.
26 Despite the tremendous progress made in understanding this process, its magnitude and
27 significance still bear large uncertainties and vary largely with aerosol distribution and
28 meteorological conditions. In this study, we particularly focus on the role of aerosol
29 vertical distribution on thermodynamic stability and PBL development by jointly using
30 the micropulse lidar, sun-photometer, and radiosonde measurements over Beijing.
31 Despite complex aerosol vertical distributions, the cloud-free aerosol structures can be
32 classified into three types: well-mixed, decreasing with height, and the inversed. Under
33 these different aerosol vertical structures, the aerosol-PBL relationships and the diurnal
34 cycles of the PBLH and $PM_{2.5}$ show distinct characters. The vertical distribution of
35 aerosol radiative forcing differs drastically with strong heating in the lower, mid, and
36 upper PBL respectively. Such a discrepancy in heating rate affects the atmospheric
37 buoyancy and stability differently in the three distinct aerosol structures. Absorbing
38 aerosol have a weak effect of stabilizing the low-atmosphere under the decreasing
39 structure than under the inverse structure. As a result, the aerosol-PBL interaction can
40 be strengthened by the inverse aerosol structure and can be potentially neutralized by
41 the decreasing structure. Moreover, aerosols can both enhance and suppress the PBL
42 stability, leading to both positive and negative feedback loops. This study attempts to
43 improve our understanding of aerosol-PBL interaction, which shows the importance of
44 the observation constraint of aerosol vertical distribution for simulating the interaction
45 and consequent feedbacks.



46 **1. Introduction**

47 Aerosols critically impact on the Earth's climate through aerosol-cloud interactions
48 (ACI) and aerosol-radiation interactions (ARI), and continue to contribute the
49 considerable uncertainty to quantifications and interpretations of the Earth's changing
50 radiation budget and hydrological cycles (Charlson et al., 1992; Ackerman et al., 2004;
51 Boucher et al., 2013; Li et al., 2011, 2017a; Guo et al., 2017; 2018). Despite the great
52 advances made in observation and modeling studies of the aerosol effects in the past
53 decades, it is still a challenge to accurately quantify the effects on the climate system
54 due to inadequate understanding of some mechanisms and strong variations in aerosol
55 type, loading, and vertical distribution (Haywood and Boucher, 2000; Carslaw et al.,
56 2013; Huang et al., 2015; Guo et al., 2016a; Li et al., 2016; Wang et al., 2019). Aerosols
57 are known to interact with thermodynamic stability through ARI (Atwater, 1971; Bond
58 et al., 2013). Absorbing aerosols can stabilize the atmosphere (Ramanathan et al., 2001;
59 Wang et al., 2013; Ding et al., 2016), whereas they may also enhance convection and
60 precipitation under certain conditions (Menon et al., 2002; Li et al., 2017).

61 Thermodynamic stability in the PBL dictates the PBL development (Garret 1994;
62 Zhang et al., 2018), thereby dominating the vertical dissipation of surface pollutants to
63 some degrees. Aerosols in turn have important feedbacks on the stability in PBL,
64 depending on the aerosol properties, especially of the light absorption aerosols (e.g.,
65 black, organic, and brown carbon). However, due to large uncertainties in aerosol
66 radiative forcing, it remains a challenge to quantify the impact of aerosols on
67 thermodynamic stability and PBL development. Conventionally, increasing the aerosol



68 absorption tends to stabilize the atmosphere, leading to reduced PBL height (PBLH). A
69 more stable atmosphere and lower PBLH will in turn increase the surface aerosol
70 loading, which is the well-established positive feedback loop in the aerosol-PBL
71 interaction (e.g., Wang et al., 2015; Ding et al., 2016; Petäjä et al., 2016; Dong et al.,
72 2017; Zou et al., 2017; Q. Huang et al., 2018; Z. Wang et al., 2018). However, such a
73 positive feedback loop may not be real for all situations and is subject to confounding
74 factors such as aerosol type, aerosol vertical distribution, soil moisture, and PBL regime
75 (Guo et al., 2019; Lou et al., 2019). Geiß et al. (2017) reported the ambiguous
76 relationships between surface aerosol loading and PBLH, while our previous study
77 revealed weak correlations between surface pollutants and the PBLH in mountainous
78 or clean regions (Su et al., 2018). A recent study by Lou et al. (2019) shows that aerosol
79 has an even positive correlation with PBLH under the stable PBL, indicating the
80 thermodynamic conditions in the PBL really matters.

81 Among others, numerical models are one of the viable methods to determine the
82 aerosol impacts on stability and PBL (e.g. Ding et al., 2016; Y. Wang et al., 2018; Zhou
83 et al., 2018; Wang et al., 2014). The aerosol optical depths (AOD), a measure of aerosol
84 columnar loading, is usually taken into account in model simulations. However, aerosol
85 vertical distribution in models is generally prescribed and may differ largely from the
86 real situation, which highly varies in the PBL and is closely linked to the significant
87 uncertainties in aerosol radiative effects. With observational constraints, the role of
88 aerosol vertical distribution in aerosol-PBL interactions warrants a further investigation.

89 Coincidentally, we have ample observational datasets over Beijing, including aerosol



90 vertical distribution derived from lidar, optical properties derived from sun-photometer,
91 the profiles of meteorological variables from radiosonde (RS), as well as surface PM_{2.5}
92 and meteorological parameters. Based on these measurements, a radiative transfer
93 model is used to simulate the vertical profiles of aerosol radiative forcing that are
94 employed to investigate the impact of aerosols on the buoyancy in the lower atmosphere.

95 The paper is structured as follows: Section 2 introduces the datasets and methods
96 used. The analyses of aerosol-PBL interaction under different aerosol vertical structures
97 are presented in Section 3. Section 5 discusses the results with a brief summary.

98

99 **2. Data and Method**

100 **2.1. Site Description**

101 In this study, we utilized data from multiple sources in Beijing, a megacity located
102 at North China Plain. As one of the most densely populated and well-urbanized regions
103 in the world, Beijing is a polluted region with high concentrations of absorbing aerosols
104 (Zhang et al., 2019). The micropulse lidar (MPL) located at Beijing was operated
105 continuously by Peking University (39.99°N, 116.31°E) from Mar 2016 to Dec 2018,
106 with a temporal resolution of 15s and a vertical resolution of 15m. Due to incomplete
107 laser pulses correction, the near-surface blind zones for lidar is ~0.15 km. Background
108 subtraction, saturation, after-pulse, overlap, and range corrections are applied to raw
109 MPL data to calculate the normalized signals (Yang et al., 2013; Su et al., 2018). The
110 MPL data on raining days are excluded. Level 1.5 AOD and single-scattering albedos
111 (SSA) are employed at multiple wavelengths (i.e. 0.44/0.5/0.67/0.87/1.02μm) from the



112 Beijing RAD1 (40°N, 116.38°E) Aerosol Robotic Network (AERONET) site during
113 2011–2018, at cloud-free conditions (Holben et al., 1998; Zhang et al., 2017; Smirnov
114 et al., 2000). The radiosonde (RS) station (39.80°N, 116.47°E) of Beijing is operated
115 by China Meteorological Administration, which is ~25 km from the MPL site. The
116 variables observed at the RS station include the meteorology data and profiles of water
117 vapor, temperature, pressure and wind. The vertical resolution of RS is altitude
118 dependent and generally less than 8 m (Guo et al., 2016b; Zhang et al., 2018). RS is
119 routinely launched at 0800 Local Time (LT) and 2000 LT for each day, and also is
120 launched at 1400 LT in summer (June-July-August). The RS measurements are
121 collected during 2011–2018. To reduce small scale bias and obtain a stable regional
122 variation of particulate matter with the diameter smaller than 2.5 μm ($\text{PM}_{2.5}$), we acquire
123 mean $\text{PM}_{2.5}$ data from twenty environmental monitoring stations within 20 km from the
124 lidar site, including one station of Beijing Embassy of United States. The topography
125 of Beijing is presented in Figure 1. The green square indicates the MPL site, and the
126 yellow triangle indicates the AERONET station. The brown star represents the
127 radiosonde (RS) station, and the red pink dots represent the $\text{PM}_{2.5}$ sites.

128 2.2. Statistical Analysis Methods

129 Here the statistical significance is tested by two independent statistical methods,
130 namely the least squares regression and the Kendall' tau test (Mann, 1945; Kendall,
131 1975). Least squares regression typically assumes a Gaussian data distribution in the
132 trend analysis, whereas the MK test is a nonparametric test without any assumed
133 functional form. The latter is more suitable for data that do not follow a certain



134 distribution. To improve the robustness of the analysis, a relationship is considered to
135 be significant when the confidence level is above 99% for both least squares regression
136 and the MK test. Hereafter, “significant” indicates the correlation is statistically
137 significant at the 99% confidence level.

138 In this study, we primarily use linear fit method to build the relationships between
139 different parameters, and the Pearson correlation coefficient derived from linear
140 regression analysis measures the degree to which the data fit a linear relationship.
141 However, following our recent work (Su et al., 2018), the inverse fitting ($f(x) = A/x +$
142 B) is used to establish the relationship between PBLH and $PM_{2.5}$. During this time, the
143 magnitude of correlation coefficient is designed to measure the degree to which the data
144 fit an inverse relationship. Since the relationship between the PBLH and $PM_{2.5}$ is non-
145 linear, the inverse fitting is more suitable to characterize this relationship.

146 2.3. PBLH and buoyancy derived from RS

147 The vertical resolution varies according to the balloon ascending rate, and RS is
148 recorded every 1.2s, which represents an approximate vertical resolution of 5–8 m.
149 Prior to the retrieval of PBLH, we further resample the radiosonde data to achieve a
150 vertical resolution of 5-hPa with linear interpolation. We follow a well-established
151 method developed by Liu and Liang (2010) to derive the PBLH based on the profiles
152 of potential temperature gradient that takes account of different stability conditions. In
153 this study, we only focus on PBL driven by buoyancy, and thus, the PBL driven by the
154 low-level jets will be excluded using the wind profiles from radiosonde (Liu and Liang,
155 2010; Miao et al., 2018).



156 The static stability in atmosphere is determined by the buoyancy force which can
157 be expressed as (Wallace and Hobbs, 2006):

$$158 \quad B = \frac{d^2z}{dt^2} = \frac{T' - T}{T} g = -g\Delta z \frac{1}{\theta} \frac{d\theta}{dz} \quad (1)$$

159 where z is the height of the air parcel and t indicates the time. T' represents the
160 temperature of the parcel and T represents the temperature of the environment and θ
161 is the virtual potential temperature of environment. For a certain layer, the atmosphere
162 is identified as a convective condition when the buoyancy is above zero, but is identified
163 as a stable condition when the buoyancy is below zero. If the buoyancy is near zero, the
164 atmosphere is under a neutral condition. Based on the identification method for PBL
165 types (Liu and Liang, 2010; Zhang et al., 2018), we present profiles of buoyancy forcing
166 for a stable, neutral, and convective PBL (Figure 2a). Clearly, the strongest upward or
167 downward forcing occurs near the surface. Figure 2b-c further show the height
168 dependent correlation coefficients between buoyancy and PBLH/PM_{2.5} with an
169 interpolation window of 0.2km. Noted the PBLH and surface PM_{2.5} are fixed for the
170 entire column, and the buoyancy is height-dependent. Due to the insufficient
171 development of PBL, we do not use RS data at 0800 LT here. To exclude the impact
172 induced by the dragging effects of rainfall, we only use the cases without precipitation
173 within the past 24 hours. Strong upward buoyancy can uplift PBLH and mitigate the
174 surface pollutants, especially in the low atmosphere. Thus, we integrate the buoyancy
175 forcing within the lowest 1km (red line in Figure 2b-c), which is defined as the low-
176 atmosphere buoyancy (LAB). As shown in Figure 3a-b, the LAB shows strong negative
177 correlations with PM_{2.5} but positive correlations with PBLH. The LAB also has



178 significant negative correlations with absorbing aerosol optical depth. It could be partly
179 caused by the stabilizing effect of absorbing aerosol on the atmosphere, which is widely
180 reported in many previous studies (Wang et al., 2015; Ding et al., 2016; Petäjä et al.,
181 2016; Dong et al., 2017; Li et al., 2017; X. Huang et al., 2018).

182 **2.4. PBLH and AEC derived from MPL**

183 MPL data from Beijing were used to retrieve the PBLH during daytime (0800-1900
184 LT). Multiple methods have been developed for retrieving the PBLH from MPL
185 measurements, such as signal threshold (Melfi et al., 1985), maximum of the signal
186 variance (Hooper and Eloranta, 1986), minimum of the signal profile derivative
187 (Flamant et al., 1997), and wavelet transform (Cohn and Angevine, 2000; Davis et al.,
188 2000; Su et al., 2017). To derive the PBLH from MPL data, we adopted previous well-
189 established approaches with several refinements, which has already been validated by
190 long-term data over Southern Great Plains (ARM SGP) site (Sawyer and Li, 2013; Su
191 et al., 2019).

192 Initially, we identify the local maximum positions (range: 0.25-4km) in the
193 covariance transform function collocated with a signal gradient larger than a certain
194 threshold. We further estimated the shot noise (σ) induced by background light and dark
195 current for each profile, and then set the certain threshold as 3σ . The initial PBLH
196 retrieval (0800LT) is constrained by the PBLH value derived from morning RS. Then,
197 the following PBLHs would be retrieved using a stability dependent model based on
198 continuity. The boundary layer clouds are identified to diagnose the PBLH for cloudy
199 cases. Figure 3d presents the comparison of summertime PBLH results derived from



200 MPL and RS at 1400 LT, and the agreement is reasonably good ($R=0.77$).

201 Multiple studies have provided a well-established algorithm to retrieve the vertical
202 profiles of aerosol extinction coefficient (AEC) from MPL (eg., Fernald, 1984; Klett,
203 1985; Liu et al., 2012). Then, the Klett method is further applied for retrieving
204 extinction profiles (Klett, 1985). The column-averaged extinction-to-backscatter ratio
205 (so-called lidar ratio) is an important parameter in the retrieval processes and is
206 constrained using AOD at $0.5\mu\text{m}$ derived from AERONET. The overall uncertainties
207 from overlap function, lidar ratio, effects of multiple scattering, and noises are
208 estimated to fall within a range of 20-30% in the retrieval processes (He et al., 2006).

209 **2.5. Estimation of the impacts of aerosols on buoyancy**

210 To illustrate the vertical profile of aerosol radiative forcing, the Santa Barbara
211 DISORT Atmospheric Radiative Transfer (SBDART) model (Ricchiazzi et al., 1998)
212 was used to simulate the atmospheric heating rate (dT/dt) induced by aerosol (Liu et
213 al., 2012; Dong et al., 2017). The integrated aerosol inputs include AODs, and SSAs
214 (i.e., 0.44, 0.67, 0.87 and $1.02\mu\text{m}$) retrieved from AERONET measurements, as well as
215 the AEC profiles at $0.5\mu\text{m}$ obtained from the MPL. We also use the MODIS surface
216 reflectance as the additional inputs
217 (<https://modis.gsfc.nasa.gov/data/dataproduct/mod09.php>). We further use heating rate
218 induced by aerosols to estimate the impacts of aerosols on buoyancy.

219 Theoretically, the rate of change in buoyancy for a certain layer can be expressed
220 as:



$$221 \quad \frac{dB}{dt} = \frac{d}{dt} \left(\frac{T_0 - \Gamma_d \Delta z - T}{T} g \right) = \frac{\left(\frac{dT_0}{dt} - \frac{dT}{dt} \right) T + \frac{dT}{dt} (\Gamma_d - \Gamma) \Delta z}{T^2} g \quad (2)$$

222 where most parameters are defined in the same way as in Eq. (1), and Γ_d (Γ)
223 represents the dry adiabatic lapse rate (environmental lapse rate). We will primarily
224 focus on the change rate in buoyancy during noontime (1100-1500LT), when PBL is
225 well developed and aerosol radiative forcing is strong. The change rate in buoyancy
226 (dB/dt) induced by aerosols is largely determined by aerosol heating rate, which can
227 be produced by the radiative transfer model. Additional inputs include environmental
228 lapse rate and temperature, which are obtained from noontime RS in the summer. For
229 other times, the environmental lapse rate and temperature are obtained from MERRA-
230 2 reanalysis data, which assimilates coarse-resolution RS observation (Rienecker et al.,
231 2011). In this way, we can estimate dB/dt induced by aerosols with a primary focus
232 on daytime. Noted the errors in MERRA-2 data would lead to the uncertainties in the
233 estimated dB/dt . 1~3 K uncertainties in MERRA-2 temperature (Gelaro et al., 2017)
234 lead to 1-3% relative biases in the estimated dB/dt . Considering the large variation in
235 dB/dt under different aerosol structures, the biases resulting from MERRA-2 data are
236 not a very serious issue.

237

238 **3. Results**

239 **3.1. Classification of different aerosol structure scenarios**

240 By altering the adiabatic heating rate of the atmosphere, the aerosol vertical
241 distribution is of great importance to the PBL. Based on cloud-free AEC profiles in the
242 PBL, aerosol vertical structures can be classified into three types: well-mixed,



243 decreasing with height, and inverse. If AEC varies by less than 20% within the lowest
244 80% of PBL, it is considered a well-mixed structure. For the other cases, a decreasing
245 structure indicates a peak in AEC near the surface, and the inverse structure indicates a
246 peak in AEC in the middle or upper PBL.

247 To investigate the vertical variation of AEC within the PBL, the evolution of PBLH
248 has been taken into account. Following previous studies (Kuang et al., 2017; Ferrero et
249 al., 2014), the vertical profiles were normalized by introducing a standardized height
250 (H_s), which was calculated as follows:

$$251 \quad H_s = \frac{z - PBLH}{PBLH} \quad (3)$$

252 where z is the height above the ground and H_s is 0 at the PBL top and -1 at ground
253 level. Then, the normalized vertical profiles of AEC during noontime are shown in
254 Figure 4. Since a temperature inversion located at the PBL top traps moisture and
255 aerosols, there is a sharp decrease in the AEC from the PBL upper boundary to free
256 atmosphere. The aerosol vertical distribution largely varies depending on different
257 conditions, but share similar features under different aerosol structure patterns. Despite
258 complex aerosol vertical distributions, these three types of profiles can account for more
259 than 85% of cloud-free cases.

260 **3.2. PBLH and PM_{2.5} under different aerosol structure scenarios**

261 Absorbing aerosols trend to have positive feedback with PBLH, while aerosols
262 vertical distribution plays a critical role in this process. We investigate the relationship
263 between MPL-derived PBLH and PM_{2.5} for absorbing ($SSA \leq 0.85$) or weakly
264 absorbing ($SSA > 0.9$) aerosols under inverse/declining aerosol structures (Figure 5).



265 In general, there are stronger correlations between PBLH and $PM_{2.5}$ under inverse
266 aerosols structure. Under such structure, the PBLH- $PM_{2.5}$ correlation also remains
267 considerably stronger for absorbing than weakly absorbing cases. This is likely caused
268 by substantial heating in the upper PBL, which would facilitate the formation of
269 temperature inversion and further increase the stability in the PBL. While, under
270 declining aerosols structure, the aerosols may not significantly redistribute the adiabatic
271 energy. This phenomenon indicates that the absorbing aerosol could play a more
272 important role in the inverse aerosol structure.

273 The diurnal cycles of AEC, PBLH, and $PM_{2.5}$ for different aerosol vertical
274 structures are presented in Figure 6 based on the measurements made in Beijing. High
275 humidity cases ($RH > 80\%$) and strong wind cases (wind speed $> 5\text{ m s}^{-1}$) are excluded.
276 Theoretically, $PM_{2.5}$ should generally decrease with increasing PBLH in the morning
277 and forenoon due to the dilution effect. This situation is demonstrated clearly for
278 decreasing structures. However, $PM_{2.5}$ continuously grows during the daytime under
279 inverse aerosol structures, regardless of the PBLH diurnal cycle. Despite the diurnal
280 variations of aerosol and PBL are controlled by many factors, the strong aerosol-
281 stability interaction may be an underlying scheme that further enhances the surface
282 aerosol loading during the daytime.

283 **3.3. Aerosol radiative forcing under different aerosol structures**

284 Figure 7 shows heating rates induced by aerosols for decreasing, well-mixed and
285 inverse cases. The vertical distributions of the heating rate differ drastically with
286 significant heating in the different parts of PBL. This is caused by substantial heating



287 in the upper PBL, which would facilitate the formation of temperature inversion and
288 further increase the stability in the PBL. Nonetheless, under the declining aerosol
289 structure, the abundant aerosols in the bottom of PBL can cause a heating effect in the
290 lower PBL, and hence, can potentially enhance the convection in PBL.

291 There are considerable differences in heating rate among the three distinct aerosol
292 structures (Figure 8), which affects the atmospheric buoyancy and stability differently.
293 On average, aerosols generally suppress buoyancy in the low atmosphere. Such an
294 effect is quite notable for inverse structure and is insignificant for decreasing structure
295 with large standard deviations. Absorbing aerosol is not very helpful for stabilizing low-
296 atmosphere under the decreasing structure, but plays an important role under the inverse
297 structure. As such, we expected the strongest aerosol-PBL interactions for absorbing
298 cases under the inverse structure, which is consistent with the results in Figure 4. It
299 should be noted that there are large variations in the impact of aerosol on buoyancy.
300 Under an inverse structure, aerosol overwhelmingly enhance the stability in low-
301 atmosphere, whereas, under decreasing structure, aerosols have the potential to either
302 enhance or suppress the low-atmosphere stability depending on different cases.

303 Figure 9 illustrates the schematic diagram of the interactions between aerosols,
304 stability, and the PBL. Overall, aerosol vertical structure critically affects the aerosol-
305 PBL interaction. The inverse aerosol structure facilitates the formation of temperature
306 inversion and further increases the stability and aerosol loading in the near surface.
307 Therefore, the inverse aerosol structure may strengthen the aerosol-PBL interaction.
308 Meanwhile, the aerosol-PBL interaction can be potentially neutralized by the



309 decreasing structure. Moreover, aerosols can both enhance and suppress the PBL
310 stability depending on different conditions, and lead to both positive and negative
311 feedback loops (Figure 9). The positive feedback loop leads to strong aerosol-PBL
312 interactions, while the negative feedback loop partly offset PBL's impacts on aerosol
313 loading. It explains the paradox of the impact of the PBL on surface pollutants, since
314 its magnitude, significance, and even sign reportedly varies or even reverses (Quan et
315 al., 2013; Tang et al., 2015; Geiß et al., 2017; Su et al., 2018).

316

317 **4. Summary and Discussions**

318 Based on integrated aerosol and meteorological measurements over Beijing, the
319 aerosol-PBL interaction is assessed under different aerosol vertical structures, which
320 are decreasing, well-mixed, and inversely increasing with height, respectively. The
321 aerosol-PBL relationships and the diurnal cycles of PBLH and $PM_{2.5}$ show distinct
322 characteristics among the different aerosol vertical patterns. For the decreasing aerosol
323 structure, $PM_{2.5}$ decreases in the morning and forenoon with relatively large PBLH
324 growth rates. In this situation, absorbing aerosol is not very helpful in stabilizing low-
325 atmosphere. Under the inverse aerosol structure, $PM_{2.5}$ continuously grows during
326 daytime with relative low PBLH growth rate. This phenomenon could be a sign of the
327 strong aerosol-PBL interaction. The aerosol radiative forcing in vertical scale for
328 decreasing, well-mixed, and inverse aerosol structures differ drastically with strong
329 heating in the lower, mid, and upper PBL respectively. Such a difference in heating rate
330 affects the atmospheric buoyancy and stability differently in the three distinct aerosol



331 structures.

332 Numerous studies used various models to simulate the aerosol-PBL interactions
333 and consequent feedbacks (e.g. Ding et al., 2016; Z. Wang et al., 2018; Zhou et al., 2018;
334 Wang et al., 2014). Aerosol vertical distribution highly varies in both temporal and
335 vertical scales, and critically affect the aerosol radiative forcing. Nonetheless, the
336 aerosol vertical distribution usually poorly represented in numerical models, due to a
337 lack of observational constraints. This study reveals the important role of aerosol
338 vertical distribution in aerosol-PBL interactions, which should be carefully taken into
339 account in both observational analysis and model simulations.

340 In this study, we use column-averaged aerosol properties from AERONET.
341 However, the vertical variations of single scattering albedo and aerosol type remains
342 unknown, which can induce uncertainties in the estimation of aerosol effects. In the
343 future, we plan to use aircraft data from field campaigns to better account for its
344 influences for different types of aerosols of different properties.

345

346 *Data availability.* The hourly PM_{2.5} data are released by the Ministry of Environmental
347 Protection of China (data link: <http://113.108.142.147:20035/emcpublish>). The
348 MERRA-2 reanalysis data are publicly available at
349 <https://disc.gsfc.nasa.gov/datasets?keywords=merra%20&page=1>. The AERONET
350 data are publicly available at <https://aeronet.gsfc.nasa.gov>. The meteorological data are
351 provided by the data center of China Meteorological Administration (data link:
352 <http://data.cma.cn/en>).



353

354 *Author contribution.* T.S. and Z.L. conceptualized this study. T.S. carried out the
355 analysis, with comments from other co-authors. C.L., J.L. and W.T. carried out the MPL
356 observation. J.G. provided auxiliary data. W.H., C.S., W.T., and J.G. provided useful
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359

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361

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369

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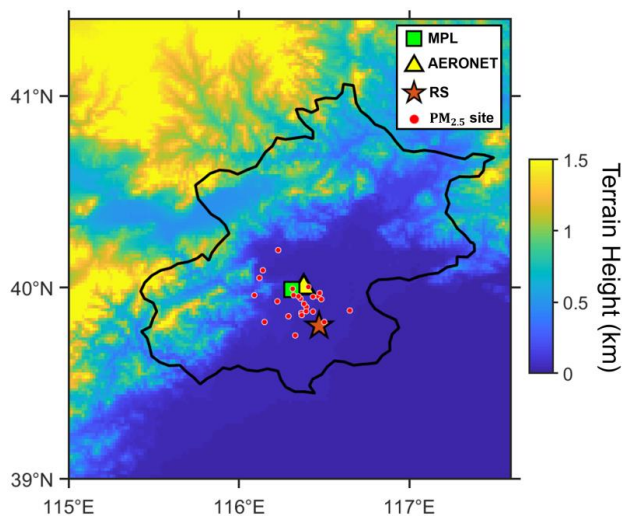
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583 **Figures**



584

585 **Figure 1.** Topography condition of Beijing. The green square indicates the MPL site,
586 and the yellow triangle indicates the AERONET station. The brown star represents
587 radiosonde (RS) station, and the red pink dots represent the PM_{2.5} sites.

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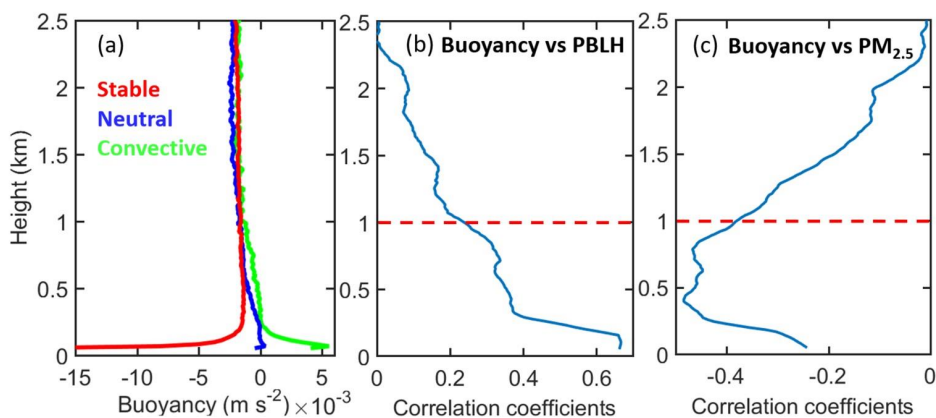
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596 **Figure 2.** (a) Vertical profiles of buoyancy forcing under stable, neutral, and convective
597 PBL. (b) Height-dependent correlation coefficients between buoyancy and PBLH. (c)
598 Height-dependent correlation coefficients between buoyancy and surface PM_{2.5}. Noted
599 the PBLH and surface PM_{2.5} are fixed for the entire column, and the buoyancy is height-
600 dependent. The buoyancy within low-atmosphere (1 km) exerts the most important
601 impact on PBLH and surface PM_{2.5}. The buoyancy and PBLH are derived from RS at
602 1400 LT and 2000 LT

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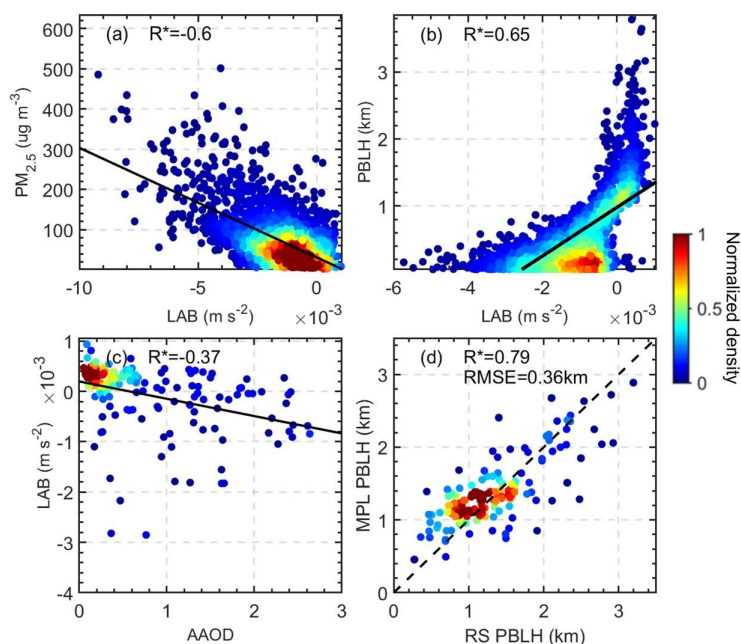
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614 **Figure 3.** (a) The relationships between low-atmosphere buoyancy (LAB) and $PM_{2.5}$.

615 (b) The relationships between LAB and PBLH. (c) The relationships between absorbing

616 aerosol optical depth (AAOD) and LAB. In (a, b, c), the LAB and PBLH are derived

617 from RS at 1400 LT and 2000 LT, and black solid lines indicate the linear regressions.

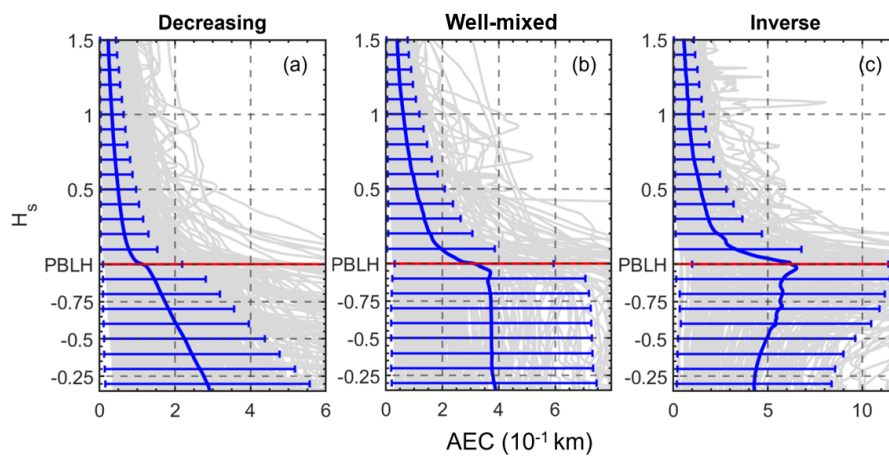
618 (d) Comparison of PBLHs derived from MPL and RS at 1400 LT. Here and in the

619 following analysis, R with asterisks indicates that the correlation is statistically

620 significant at the 99% confidence level. The color-shaded dots indicate the normalized

621 sample density.

622



623

624 **Figure 4.** The normalized vertical profiles of aerosol extinction coefficients (AEC)
625 under (a) decreasing, (b) well-mixed, and (c) inverse structures. The red line marks the
626 position of the PBLH, the solid blue lines represent the average profile of corresponding
627 profiles, and the error-bars represent the standard deviations.

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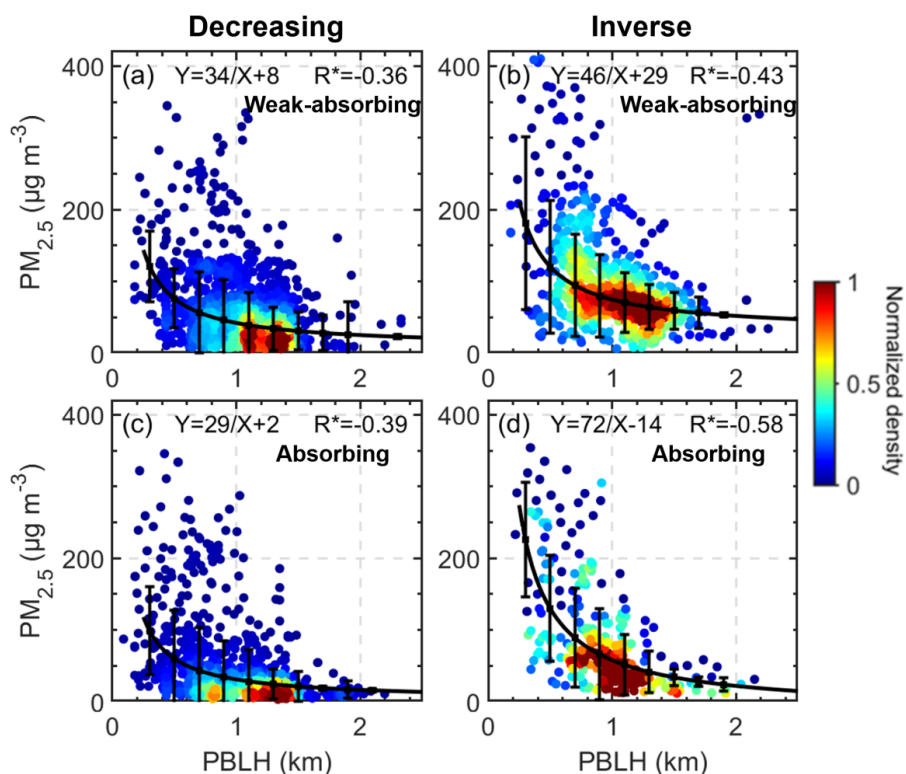
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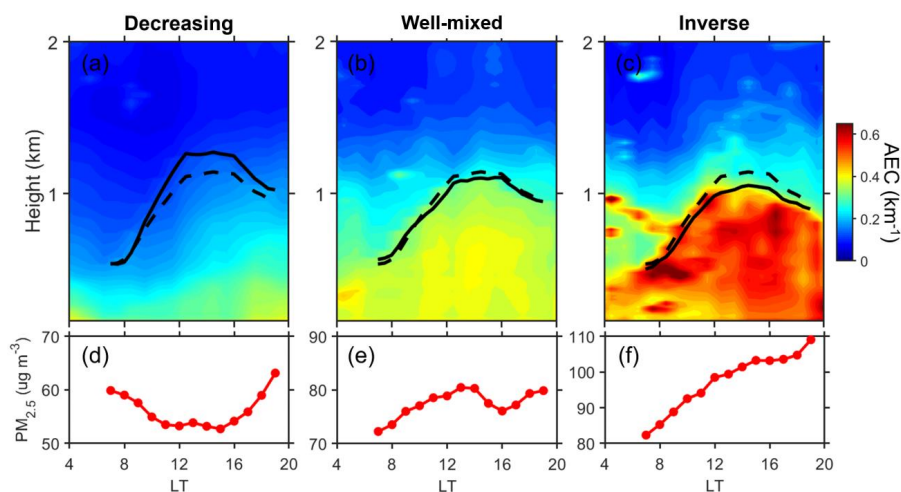


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635 **Figure 5.** The relationship between MPL-derived PBLH and $PM_{2.5}$ for (a) weak-
636 absorbing and (c) absorbing under decreasing aerosols structure. The relationship
637 between MPL-derived PBLH and $PM_{2.5}$ for (b) weak-absorbing and (d) absorbing under
638 inverse aerosols structure. The black lines represent the inverse fit, and the whiskers
639 indicate the standard deviations. The detailed fitting functions are given at the top of
640 each panel, along with the correlation coefficient for the inverse fit.

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644 **Figure 6.** Diurnal variations in AEC for (a) decreasing, (b) well-mixed, and (c) inverse
645 structures. The solid black lines indicate the corresponding PBLH diurnal cycles. The
646 dashed black line represents the mean PBLH diurnal cycle. (d, e, f) The corresponding
647 diurnal variations in PM_{2.5}.

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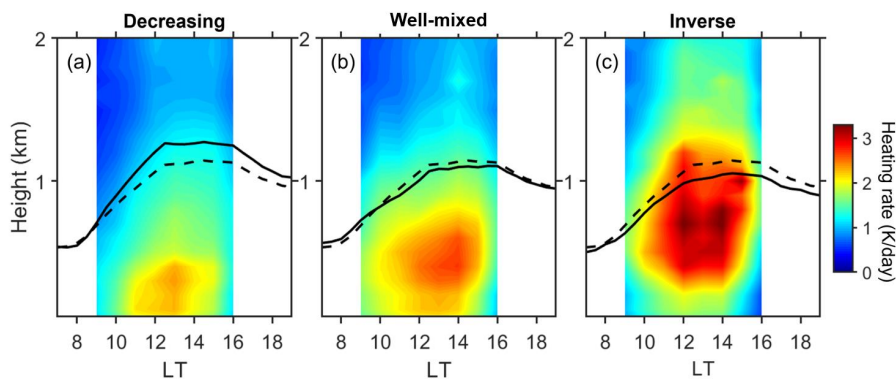
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658 **Figure 7.** The profiles of aerosol radiative forcing for (a) decreasing, (b) well-mixed,
659 and (c) inverse structures of aerosol loading. The solid black lines indicate the
660 corresponding PBLH diurnal cycles. The dashed black line represents the mean PBLH
661 diurnal cycle.

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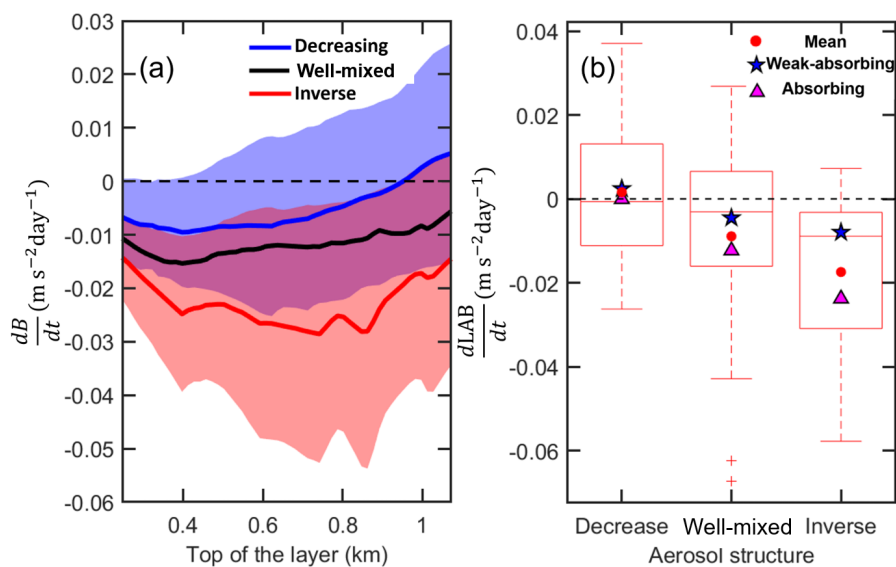
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671 **Figure 8.** (a) The change rate in buoyancy ($\frac{dB}{dt}$) in a certain layer of lowest
672 atmosphere under decreasing (blue), well-mixed (black), and inverse (red) aerosol
673 structures during noontime. The change rate at the bottom of the layer is zero, and that
674 in buoyancy is subjected to the top of the layer. The shaded areas show the standard
675 deviation of change rate in buoyancy. (b) Box-and-whisker plots showing 10th, 25th,
676 50th, 75th, and 90th percentile values of change rate in LAB (buoyancy within lowest
677 1km) during noontime. The red dots indicate the mean values, while the blue stars and
678 pink triangles show means for weak-absorbing ($SSA > 0.9$) and absorbing ($SSA < 0.85$)
679 cases.

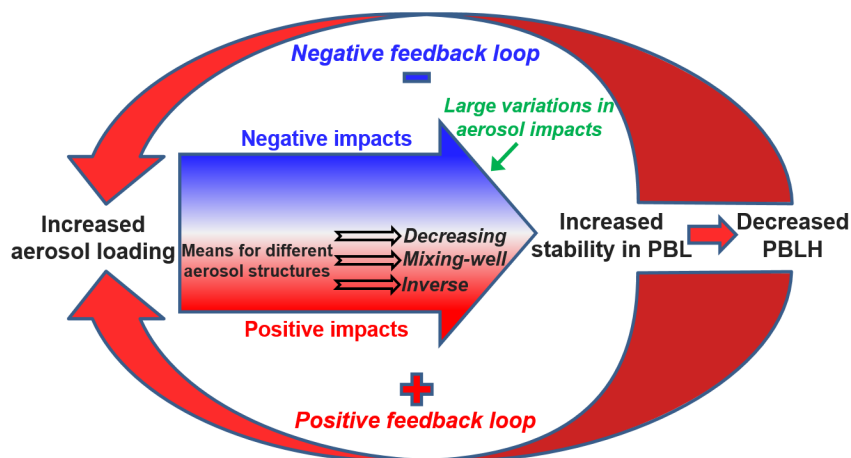
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686 **Figure 9.** Schematic diagram showing the interactions between aerosols, stability, and
687 the PBL that comprise positive impacts (red arrows) and negative impacts (blue arrows).
688 The aerosol impacts on stability largely vary with meteorology and aerosol conditions,
689 and the means of aerosol impacts on stability for different aerosol structures are
690 indicated in the diagram.

691